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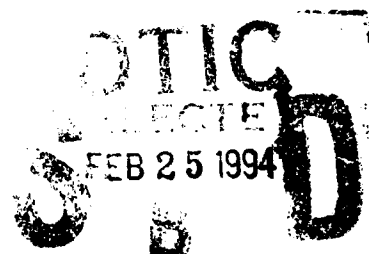
**NRL Memorandum Report 6649**

# **The Stability and Structure of Lean Hydrogen-Air Flames: Effects of Gravity**

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13. ABSTRACT (Maximum 200 words) Detailed, time-dependent, two-dimensional numerical simulations with full hydrogen-oxygen chemistry are used to investigate the effects of gravity on the stability and structure of laminar flames in lean, premixed hydrogen - air mixtures. The calculations show that the effects of gravity becomes more important as the lean flammability limit is approached. In a 12% hydrogen - air mixture, gravity plays only a secondary role in determining the multidimensional structure of the flame with the stability and structure of the flame controlled primarily by the thermo-diffusive instability mechanism. However, in leaner hydrogen-air mixtures gravity becomes more important. Upward-propagating flames are highly curved and evolve into a bubble rising upwards in the tube. Downward-propagating flames are flat or even oscillate between structures with concave and convex curvatures. The zero-gravity flame shows only cellular structures. Cellular structures which are present in zero gravity can be suppressed by the effect of buoyancy for mixtures leaner than 11% hydrogen. These observations are explained on the basis of an interaction between the processes leading to buoyancy-induced Rayleigh-Taylor instability and the thermo-diffusive instability.				
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# THE STABILITY AND STRUCTURE OF LEAN HYDROGEN-AIR FLAMES: EFFECTS OF GRAVITY

## Introduction

Multi-dimensional structures are often observed in propagating flames, especially near the flammability limits. These structures may arise due to various types of instabilities that can occur in such premixed flames. In this paper, we examine these instabilities and use numerical simulations to isolate and study their effect on the flame structure. The emphasis of this paper is on the effects of gravity on flame instabilities and structure in gases of premixed hydrogen and air.

Linear stability analyses can provide information on the roles of various processes at the onset of instability. However, the prediction of the growth of this instability to the final form is beyond the scope of these analyses. Numerical calculations can be used to help understand both the onset of the instability and the evolutionary process that produces the multidimensional structure. The numerical simulations of flames presented here include as input a multireaction mechanism for hydrogen combustion, molecular diffusion between the reactants, intermediates, and products, thermal conduction, convection, and gravity. Such a detailed model allows us to investigate the multidimensional structure of flames and to evaluate the relative importance of various instability mechanisms in normal earth gravity and zero-gravity conditions.

The three major instabilities that might occur in premixed flames are: the hydrodynamic instability, independently proposed by Landau<sup>1</sup> and Darrieus<sup>2</sup>; the thermo-diffusive instability investigated by Barenblatt<sup>3</sup> and Zeldovich<sup>4</sup>; and the buoyancy-induced instability, generally called the Rayleigh-Taylor instability<sup>5</sup>. A numerical model of the premixed hydrogen flame that includes all the physical mechanisms that lead to these three types of instabilities provides an ideal test-bed for studying these instabilities.

Hydrodynamic instability was the first kind of flame instability studied theoretically. The analyses of Landau<sup>1</sup> and Darrieus<sup>2</sup> showed that a planar flame, considered as a density discontinuity that propagates at a constant speed, is unstable to wavelengths of all sizes. Hydrodynamic instabilities can be expected to occur in all flames in the absence of stabilizing mechanisms. The analysis by Markstein<sup>6</sup> has shown that introducing a characteristic length of the order of the flame thickness has a stabilizing effect on the short wavelength modes. This instability is expected to become important in large systems which are much larger than the flame thickness. In the studies presented here the hydrodynamic instability is not expected to be important because of the relatively small system size.

The thermo-diffusive instability mechanism proposed by Zeldovich<sup>4</sup>, Barenblatt et al.<sup>3</sup>, and Sivashinsky<sup>7,8</sup> involves a competition between mass diffusion of the deficient reactant and diffusion of heat in the mixture. For a simple one-step reaction in an effectively single-reactant system, this mechanism predicts the formation of cellular structure whenever the thermal diffusivity of the mixture is sufficiently smaller than the mass diffusivity of the reactant. For lean hydrogen-air mixtures, as examined in this study, hydrogen is the deficient reactant and its mass diffusivity significantly exceeds the thermal diffusivity of the mixture. In rich hydrogen-air mixtures, oxygen is the deficient reactant and its mass diffusivity is nearly the same as the mixture thermal diffusivity. Hence this theory agrees with early experimental observations<sup>9,10</sup> of unstable lean hydrogen-air mixtures and stable rich mixtures. More recent experiments<sup>11,12</sup> have shown cellular flames in some rich and near-stoichiometric hydrogen-air mixtures, contradicting this simple theory. The theory has been extended<sup>13,14</sup> to near-stoichiometric mixtures by considering both the deficient and abundant components.

The Rayleigh-Taylor instability occurs when a heavier fluid is accelerated into a lighter fluid. On earth, this acceleration is provided by gravitational attraction. In an upward-propagating flame, the light, hot burned material is on the bottom, and the dense, cold unburned material is on the top, resulting in instability. In a downward-propagating flame, the light material is on the top, and the Rayleigh-Taylor mechanism stabilizes the system. The physical mechanisms causing the thermo-diffusive instability and this buoyancy-induced instability can be important simultaneously so that under certain conditions, this interaction appears to suppress the formation of cellular structure<sup>15</sup>. Dimensional arguments<sup>16</sup> and theoretical analysis<sup>17</sup> indicate that the importance of the buoyancy-induced instability increases as the flame speed decreases, and hence is more important when the mixture is near its flammability limits.

Cellular structures in flames have been observed in the microgravity experiments in the NASA drop tower<sup>18</sup> and Lear-jets<sup>19</sup>. These flames in microgravity are essentially free from any buoyancy-induced instability.

Our numerical simulations of premixed flames contain detailed models of the physical processes that cause the various instabilities. Previously we have used such simulations to show that, in the absence of gravity, cellular structure is caused by the thermo-diffusive mechanism<sup>20</sup>. We have shown that this mechanism is correct for mixtures sufficiently far from stoichiometric. In this paper, we investigate the effect of gravity on flame instability and, in particular, focus on the interaction between the processes leading to thermo-diffusive instability and the Rayleigh-Taylor instability and their effect on flame structure.

## Multidimensional Flame Model

A detailed model of a flame must contain accurate representations of the convective, diffusive, and chemical processes. The individual importance of these processes varies from rich to lean flames, and is especially notable near the flammability limits<sup>21</sup> where the exact behavior of these flames depends on a delicate balance among the processes. The reactive-flow conservation equations are solved for density, momentum, energy and the species number densities. In the rest of this section, we briefly describe the algorithms and input data used to model and couple the various physical processes. Full details of the numerical scheme can be found in Reference 22.

The fluid convection algorithm must be able to maintain the sharp gradients present in flames. This means that the numerical diffusion in the calculation must be considerably less than any important physical diffusion. The BIC-FCT algorithm<sup>23</sup> was developed specifically to solve low-speed flow problems with high accuracy. BIC-FCT combines an explicit high-order, nonlinear FCT method<sup>24,25</sup> with an implicit correction process. This combination maintains monotonicity and high accuracy but removes the timestep limit imposed by the speed of sound. By using BIC-FCT, spurious numerical oscillations that would lead to unphysical chemical reactions do not occur.

Thermal conductivity of the individual species is modeled by a polynomial fit in temperature to existing experimental data. Individual conductivities are then averaged using a mixture rule<sup>26,27</sup> to get the mixture thermal conductivity. A similar process is used to obtain the mixture viscosity from individual viscosities. Heat and momentum diffusion are then calculated explicitly using these coefficients. All viscous terms in the compressible

Navier-Stokes equation have been included.

Binary mass diffusion coefficients are represented by an exponential fit to experimental data, and the individual species diffusion coefficients are obtained by applying mixture rules<sup>26</sup>. The individual species diffusion velocities are solved for explicitly by applying Fick's law followed by a correction procedure to ensure zero net flux<sup>27</sup>.

Chemistry of the hydrogen-oxygen flame is modelled by a set of 24 reversible reaction rates describing the interaction of eight species,  $H_2$ ,  $O_2$ ,  $H$ ,  $O$ ,  $OH$ ,  $HO_2$ ,  $H_2O_2$ ,  $H_2O$ , and  $N_2$  is considered a nonreacting diluent<sup>28</sup>. This reaction set is solved at each timestep with TBA, a vectorized version of CHEMEQ, an integrator for stiff ordinary differential equations<sup>29</sup>.

All the chemical and physical processes are solved sequentially and then are coupled asymptotically by timestep splitting<sup>30</sup>. This modular approach greatly simplifies the model and makes it easier to test and change the model. Individual modules were tested against known analytic and other previously verified numerical solutions. One-dimensional predictions of the complete model were compared to those from the Lagrangian model FLAME1D which has been benchmarked extensively against theory and experiment<sup>25</sup>.

## Results and Discussion

Initial conditions for the two-dimensional calculations were obtained by performing a one-dimensional calculation to provide the conditions for steady, propagating flames. Figure 1 shows the configuration under study and gives the boundary conditions of the computational domain. Fresh unburned gas flows in from the left, and the products of chemical reaction at the flame front flow out to the right. If the inlet velocity is set to the burning velocity of the flame, the flame zone is fixed in space and there is a steady, propagating flame. Thus, the transient effects arising from the ignition process can be eliminated and the one-dimensional solution provides the initial condition for the two-dimensional calculation. The computational domain for the two-dimensional calculation was  $5.1 \text{ cm} \times 12 \text{ cm}$ , which was resolved by a  $144 \times 128$  variably spaced grid. Fine zones were clustered around the flame front. Calculations have been performed for several  $H_2$  - air mixtures, ranging from 15% hydrogen down to 7.5% hydrogen. However, only calculations for 12, 11, and 10% hydrogen-air mixtures will be presented in detail.

### 12% Hydrogen - Air Flames

The first series of calculations described here are of flames in a fuel-lean mixture of 12% hydrogen in air. Flames propagating upward and downward in normal gravity and flames

propagating in a zero-gravity environment are simulated. In all cases, the initial condition described by Fig. 1 is perturbed by displacing the center portion of the planar flame against the direction of the flow. In each case, the effect of the disturbance on the planar flame and the subsequent evolution of a multidimensional structure is studied in detail and compared in Fig. 2. This figure presents the OH radical concentrations for upward-, zero-gravity, and downward-propagating flames at a sequence of times. For the three cases in this figure (and in Figs. 3 and 5), the sequence of frames at various times are aligned such that time advances from the bottom to the top and the fresh, unburned mixture flows in from the top in each case. This convention in presentation has been adopted for ease in comparing the relative rate of changes in the three cases although this results in the apparent reversal of the direction of propagation of the downward-propagating flame.

The first observation we make from this figure is that in all three cases, the planar flame is unstable and evolves into multidimensional structures. At first glance, all three flames are similar and exhibit the characteristic features of cellular flames. To highlight the differences due to the effect of gravity, it is helpful to examine the shapes of the flames at two different length scales. The longer length scale is on the order of the size of the channel while the smaller length scale is on the order of the thickness of the flame. Using this perspective, the multidimensional structure of the flame can be described as a large-scale curvature of the flame front on which is superposed small scale cell-like structures. These small cells are due to the thermo-diffusive instability and are present in any gravity, independent of the direction of propagation. The shape of these small structures and the size of these cells appear to be largely unaffected by gravity.

However, there is some effect of gravity on the overall large-scale shape of the flame. In the upward propagating flame, where the buoyancy forces are also destabilising in addition to the thermo-diffusive instability mechanism, the flame has a more curved shape than the zero-gravity flame. The downward-propagating flame has a flatter overall shape, with the large-scale curvature of the flame suppressed by the stabilizing effect of buoyancy. The most pronounced effect of gravity can be seen between 37.5 and 45 ms. During this period, the upward propagating flame undergoes a bubble-like growth which enhances the overall curvature of the flame. For the downward propagating flame, it is mainly during this period that the flame front flattens out.

An interesting feature of this mixture is that the cells which form grow and then split into two. These newly formed cells grow and subsequently split. This indicates that there is

a preferred size for the cells, as has been observed in experiments. This subsequent splitting had not been observed in our earlier calculations.

The fact that a planar flame is unstable in all three cases and evolves to a cellular structure is consistent with our previous results<sup>20</sup> that show that cellular structure is due to a thermo-diffusive instability mechanism. Our current calculations show that in this mixture, the effect of buoyancy through the Rayleigh-Taylor instability is not substantial. The dimensional arguments presented by Williams<sup>16</sup> and the theoretical analysis of Clavin<sup>17</sup> indicate that the buoyancy-induced instability becomes more important as the flame speed decreases. Therefore, we have performed computations for still leaner mixtures which have lower burning velocities.

### **11% Hydrogen - Air Flames**

As expected, the 11% mixture exhibits a stronger effect of gravity, primarily on the overall large-scale shape of the flame and to a lesser extent on the evolution of the small scale cell-like structures. This can be seen in Fig. 3, in which the OH radical concentrations for upward-, zero-gravity, and downward-propagating flames are shown.

The upward propagating flame is highly curved and by 100 ms attains the appearance of a large, corrugated bubble rising up the channel. The zero-gravity flame is much less curved, without the bubble-like formation. The downward propagating flame has a flat overall shape throughout the calculation. For this mixture, the effect of gravity on the flame shape becomes apparent by 60 ms and thereafter controls the shape of the flame. The small scale cell-like structures are still present in all gravities. These cellular structures attain a larger size than in the 12% mixture prior to splitting. This can be most clearly seen in the flame in zero gravity. The large central cell which has been present in all the cases discussed so far is suppressed in the downward propagating flame. The effect of gravity on the cellular structures is observed to be significant for this mixture.

Downward propagating flames are of special interest because their cellular structures have been investigated experimentally<sup>11</sup>. Therefore we have extended the calculations for this case up to 160 ms, as shown in Fig. 4. The flame is seen to be flat, with distinct cellular structures. These cellular structures split in two when they grow beyond a critical size. The newly created cells grow and split in turn. Thus, a preferred cell size is established.

### **10% Hydrogen - Air Flames**

Figure 5 shows the OH contours for 10% hydrogen - air flames. Results for this mixture

are very similar for both the upward and zero-gravity flames as in the 11% mixture. However, the evolution of the structures is slower, with the effect of gravity becoming dominant later, at 80 ms. The evolution of the cellular structures and their splitting in zero gravity is also delayed, with the structure at 140 ms resembling that of 80 ms in the 11% mixture. Other than the delay, both the large-scale and small-scale structures that evolve are quite similar.

However, a marked difference can be seen in the downward propagating flame. In this mixture, no small cells are formed, and the flame is quite smooth. The initial central cell is completely suppressed by the stabilizing effect of buoyancy, and unlike the less lean mixtures, no new cells are formed. The large scale structure of the flame undergoes a slow oscillation about its planar position. Extended calculations show that this oscillation is damping out. For the downward propagating flames in this mixture and in still leaner mixtures, the structures caused by the thermo-diffusive instability are overwhelmed by those arising from buoyancy. Any cellular structures which form early are overtaken by buoyancy effects with the result that the upward-propagating flame evolves into a bubble-like surface rising in the channel and the downward-propagating flame oscillating between mildly concave and convex flame shapes.

## Conclusions

Detailed two-dimensional numerical simulations of flame instabilities in lean hydrogen - air mixtures have been carried out for upward and downward propagating and zero-gravity flames near the flammability limit. Physical processes included in the model are: fluid convection, detailed hydrogen-oxygen chemistry, multi-species diffusion, thermal conduction, viscosity, and gravity. The simulations show the characteristic cellular structure observed in experiments and predicted by theory. The thermo-diffusive instability was found to be present in the mixtures studied and at both orientations of gravity and is responsible for the small, cell-like structures. However, presence of the buoyancy-induced instability alters the large scale structure of the flame. The effect of buoyancy on the cellular structures formed by the thermo-diffusive instability is small in mixtures which have greater than 11% hydrogen. In a 11% mixture, a downward propagating flame is flat with small cells that repeatedly split. In leaner mixtures, the effect of gravity is more dramatic. In these mixtures, the upward propagating flame had the characteristic bubble shape observed experimentally<sup>31</sup> and the downward propagating flame had oscillations characteristic of the Rayleigh-Taylor instability and does not have any cellular structures. These results agree with the theory<sup>17</sup>

that indicates that the influence of gravity is greater for lower flame speeds and that such oscillatory behavior is possible. These results indicate that the instability mechanisms can interact in a quite complex manner, and even though one mechanism can mask the other, in certain regimes, both can be equally important.

Calculations for still leaner mixtures are needed to address the actual extinction behavior of upward and downward-propagating flames. Loss mechanisms such as heat and radical losses to the walls as well as radiation might also play a role in determining the detailed extinction behavior of these flames. These effects will be systematically considered in further calculations.

### **Acknowledgements**

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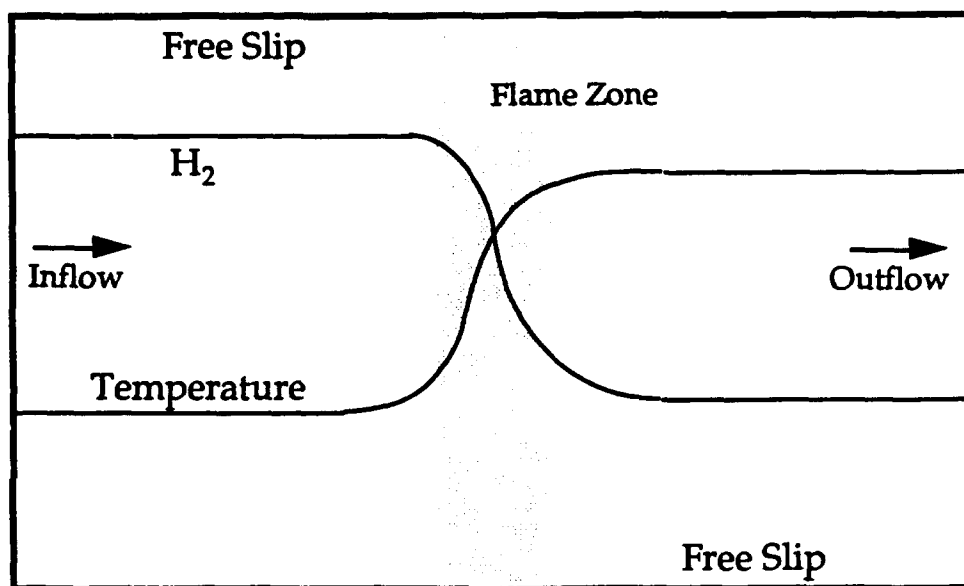


Figure 1. Initial and boundary conditions for the two-dimensional flame calculations.

12% H<sub>2</sub> - Air Mixture  
OH Concentration

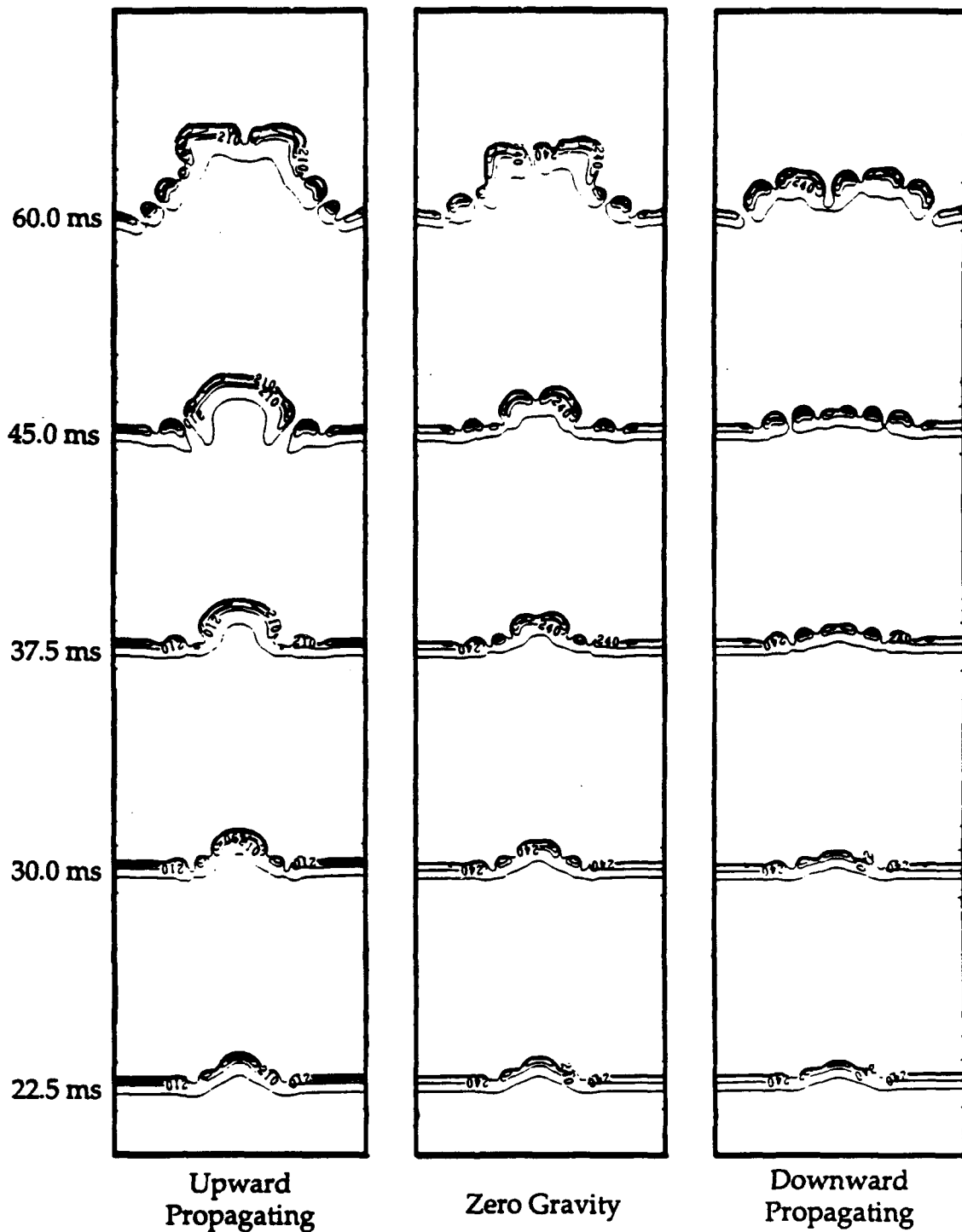


Figure 2. OH radical number density contours for flames in a 12% H<sub>2</sub> - air mixture, contour levels scaled by  $10^{-14}$ .

11% H<sub>2</sub> - Air Mixture  
OH Concentration

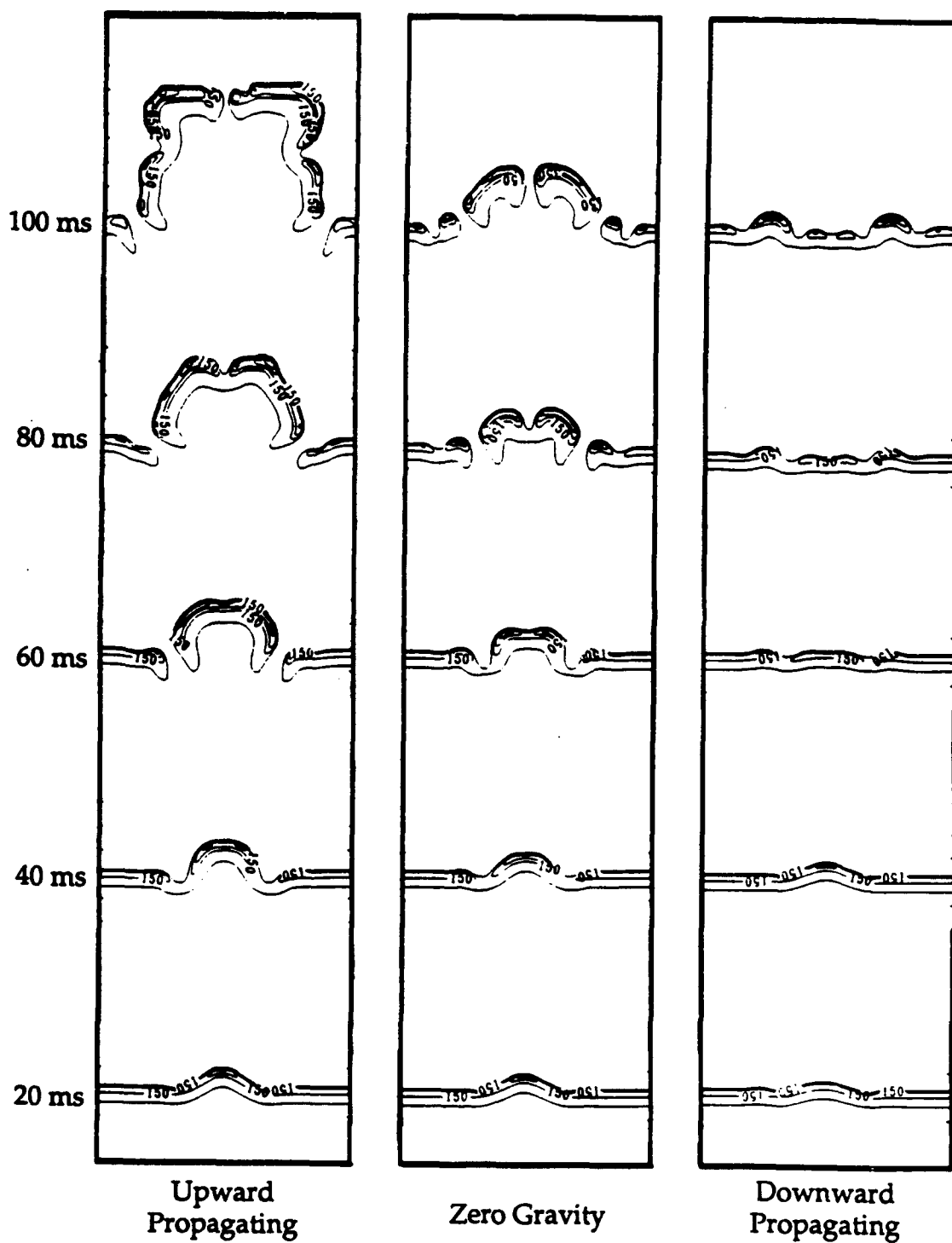


Figure 3. OH radical number density contours for flames in a 11% H<sub>2</sub> - air mixture, contour levels scaled by  $10^{-14}$ .

11% H<sub>2</sub> - Air Mixture  
OH Concentration, Downward Propagation

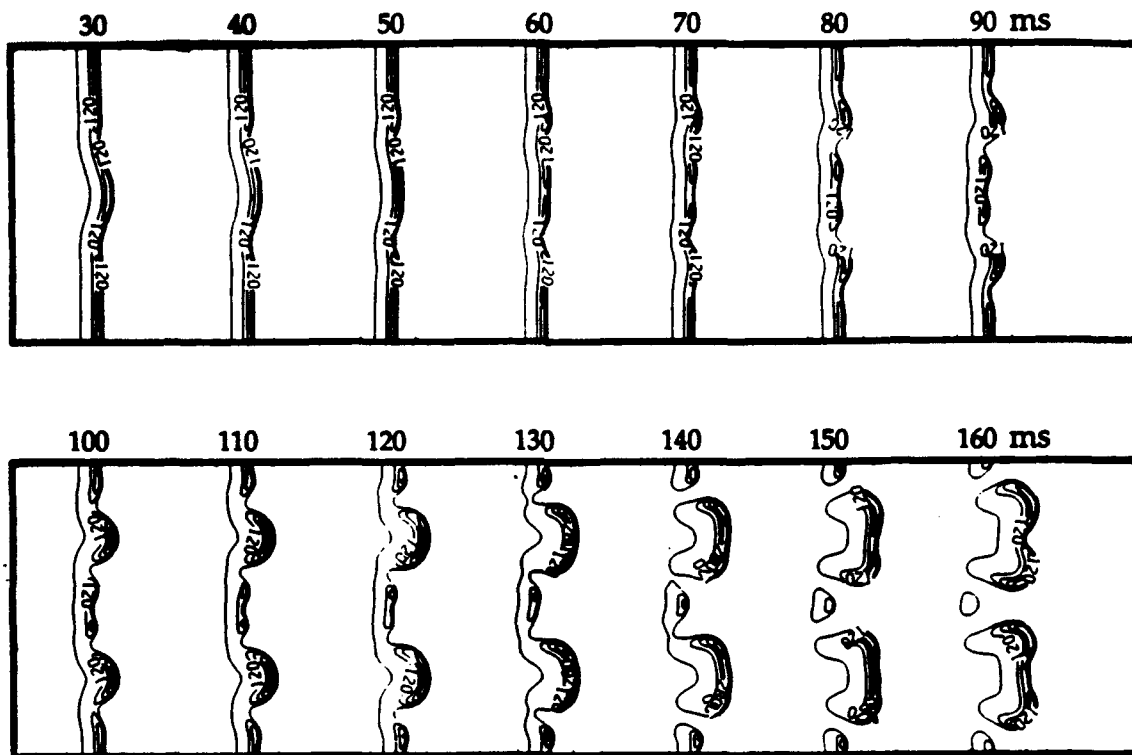


Figure 4. OH radical number density contours for a downward propagating flame in a 11% H<sub>2</sub> - air mixture, contour levels scaled by  $10^{-14}$ .

10% H<sub>2</sub> - Air Mixture  
OH Concentration

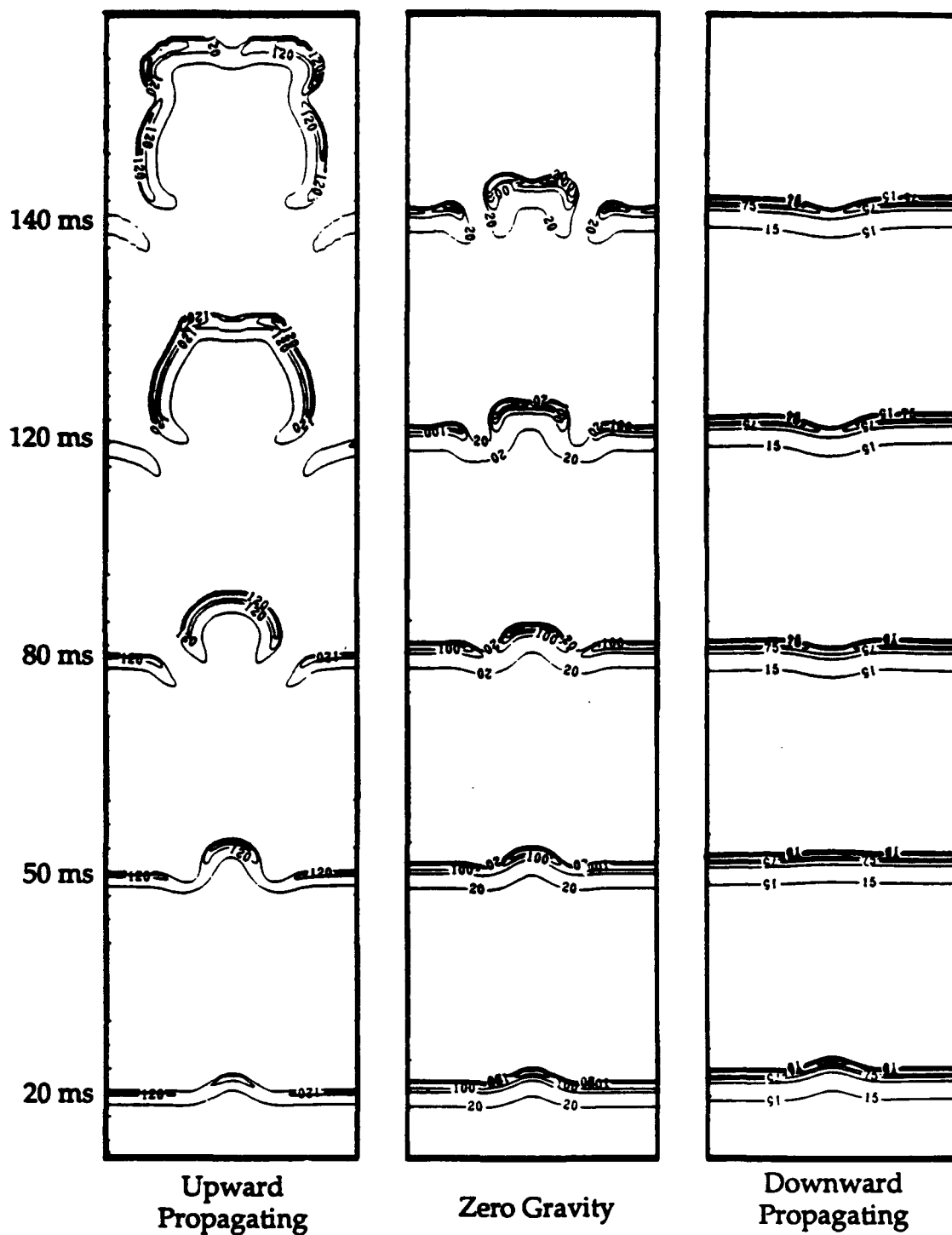


Figure 5. OH radical number density contours for flames in a 10% H<sub>2</sub> - air mixture, contour levels scaled by  $10^{-14}$ .